Supplementary Information for "Impact of Rheology on Formation of Oil-in-Liquid Metal Emulsions"

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S1. Rheological measurement procedure

We used a TA Instrument Discovery Series Hybrid (DHR-2) Rheometer equipped with nitrogen gas purge and environmental chamber to control temperature. We conducted the measurements using a 25 mm parallel plate geometry attachment in sweeping mode while heating the sample to 45 °C. After about 0.5 mL of the sample was applied to the bottom plate, the top plate was lowered onto the sample to a final thickness of around 300 µm and a compressive force under 5 N (in the process of lowering the plate, part of the LM leaks out decreasing the measured volume to about 0.15 mL). The excess sample was removed using the trimming tool from the parallel base plate.

The viscosity of the Ag-in-Ga pastes were measured on the same day as the samples were fabricated, with sufficient time provided for alloying to occur (as confirmed with imaging and presence of only nanoneedle particles). We note that in prior literature it was demonstrated that the alloying process is very rapid,[10,11,30–32] therefore even a relatively short period post fabrication is sufficient for the samples to be finalized (~2 h). The measurements were done on 3 separate days; wherein, on each day, we did only 1 sample for each concentration between 0-2.5 wt.% . All samples were kept under static vacuum (for inert environment) and 45 °C in vacuum ovens until it was tested on the rheometer. The sample were kept in this environment to prevent reactions between external gallium oxide shells with moisture in environment, which can occur during contact at elevated temperatures over longer periods of time. To test 1 sample at shear rates between 0.1 - 100 s^{-1}, it approximately took 560 seconds and the step time at each shear rate was 35 s.

S2. Viscosity for the top layer of LM-oxide foams for different times of mixing

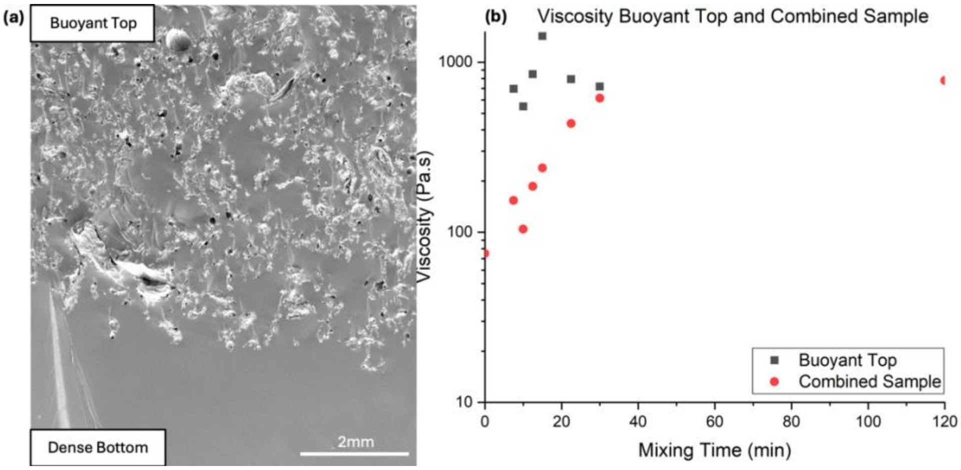

Figure S1. Represents (a) an electron micrograph demonstrating the segregation of the less dense top and more dense bottom part of LM-oxide foams. (b) The plot demonstrates the viscosities (measured at 1 s^{-1}) for the buoyant region and combined sample for different mixing times at 600 rpm. The top layer was separated from the bottom through sample tilting into a separate container.

S3. Viscosity measurements of all the Ag-in-Ga samples

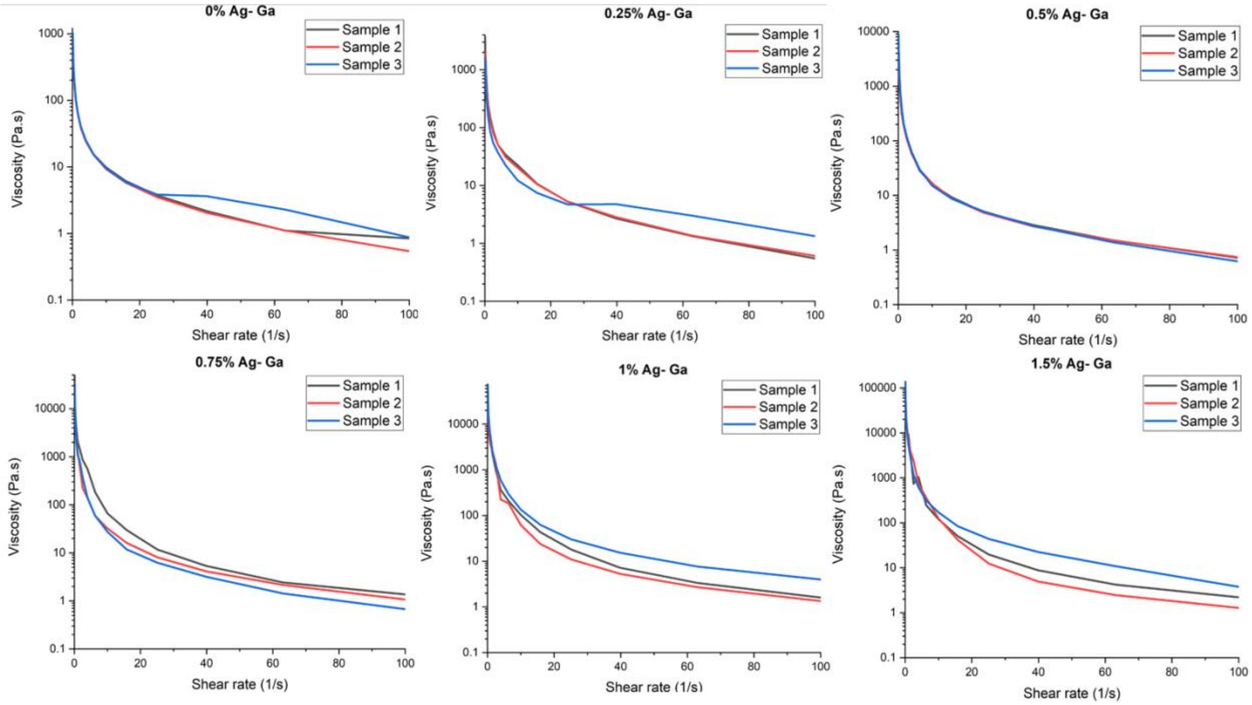

Figure S2. Viscosity as function of shear rates for each volume percentage of Ag in Ga with measurements for three separate samples shown.

S4. Viscosity of the silicone oils

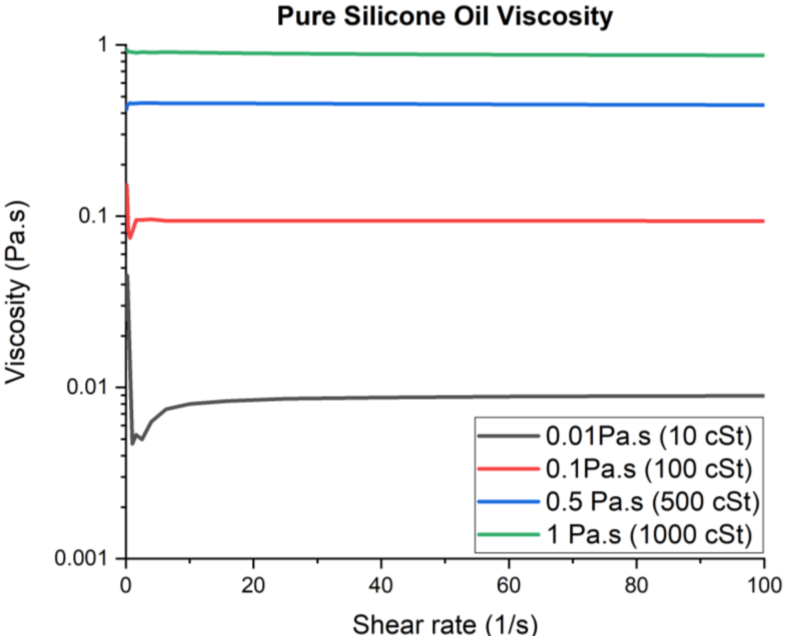

Figure S3. Viscosity as a function of shear rates for the utilized silicone oils.

S5. Environmental Scanning Electron Microscopy of the samples

The frozen samples of Ag-in-Ga mixtures and SO-in-LM emulsions were cut as in our prior work and imaged using Thermo Fisher Scientific Prisma Environmental Scanning Electron Microscope with thermal emission source used in low vacuum mode with Low Vacuum Detector, 1 Torr of water vapor chamber pressure, beam voltage of 20 kV and 1.2 to 17 nA of current. The presence of the water vapor facilitates imaging of partially electrically insulating samples such as silicone oil. We found that image quality was substantially improved by lightly passing a Kim wipe across the cross-sectioned surfaces aided in removal of excess silicone oil that smeared over the freshly cleaved surfaces.

S6. ESEM of silver particles

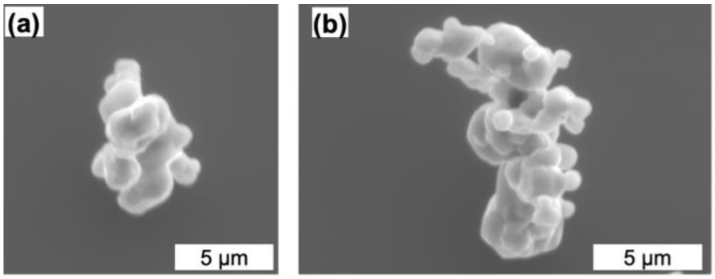

Figure S4. Electron micrographs of the silver particles prior to mixing and alloying with Ga.